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**A CALIBRATION OF THE PRODUCTION RATE RATIO  $P_{21}/P_{26}$  BY LOW ENERGY SECONDARY NEUTRONS: IDENTIFICATION OF Ne SPALLATION COMPONENTS AT THE  $10^6$  ATOMS/g LEVEL IN TERRESTRIAL SAMPLES.** Th. Graf, S. Niedermann<sup>1</sup> and K. Marti, Dept. of Chemistry, Univ. of California, San Diego, La Jolla, CA 92093-0317.

The spallation ratio  $(^{22}\text{Ne}/^{21}\text{Ne})_c$  from Si was determined as  $1.243 \pm 0.022$  in a terrestrial quartz sample. We carried out a calibration of the *in-situ* production rate ratio  $P_{21}/P_{26}$  in quartz samples for which  $^{10}\text{Be}$  and  $^{26}\text{Al}$  production rates were previously measured. A ratio  $P_{21}/P_{26}$  of  $0.67 \pm 0.12$  is obtained.

Recent improvements in experimental techniques make it possible to measure a number of *in-situ* produced cosmogenic radionuclides as well as  $^3\text{He}$  and  $^{21}\text{Ne}$  in terrestrial rock samples. Unfortunately, the potentially important applications in geomorphology and glaciology are at present limited because the production rates of stable  $^{21}\text{Ne}$  are poorly known. This study attempts to remedy the situation by first calibrating the spallation ratio  $(^{22}\text{Ne}/^{21}\text{Ne})_c$  required for identification of *in-situ* produced cosmogenic components, and second calibrate the production rate ratio  $P_{21}/P_{26}$ .

We analyzed several splits of a quartz separate from Antarctic sandstone boulder ALH85-4 (collected in 1882m altitude in the Mawson formation). Quartz was separated according to [1].  $^{10}\text{Be}$  and  $^{26}\text{Al}$  results [2] yield a minimum exposure age of  $1.06 \pm 0.27$  Ma and a maximum erosion rate of  $0.36 \pm 0.05$  m  $\text{Ma}^{-1}$ . The long exposure time accounts for a significant amount of cosmic-ray-produced  $^{21}\text{Ne}$  ( $\approx 150 \times 10^6$  atoms/g), which is expected to outweigh any nucleogenic components in this rock. Fig. 1 shows that data from the four aliquots lie on a straight line passing through the "AIR" point, within error limits. The simplest interpretation is that Ne in ALH85-4 is a two component mixture of trapped Ne ( $\text{Ne}_{tr}$ ) with essentially atmospheric composition and cosmic-ray produced Ne.  $^4\text{He}$  can be used to constrain a possible superposition of a Ne component produced by the reaction  $^{18}\text{O}(\alpha, n)^{21}\text{Ne}$ : Assuming that at least 1% of  $^4\text{He}$  is retained in the sample, less than 5% of the  $^{21}\text{Ne}$  excess can be of nucleogenic origin. The slope of the correlation line in Fig. 1 is  $1.120 \pm 0.021$ . For a production ratio  $(^{21}\text{Ne}/^{20}\text{Ne})_c = 0.8 \pm 0.1$  adopted for Si, we obtain a ratio  $(^{22}\text{Ne}/^{21}\text{Ne})_c = 1.243 \pm 0.022$ . Slightly higher ratios of 1.3-1.4 were calculated by Michel et al. [3] and Hohenberg et al. [4] for meteorites and the lunar surface, respectively. This ratio should be independent of latitude and altitude of the sampling site, because the cosmic-ray energy spectrum becomes invariant at (atmospheric) depths  $>200\text{g}/\text{cm}^2$ , and negative muon captures contribute little to the Ne production from Si.

$^{21}\text{Ne}_c$  is released from quartz at very low temperatures. More than 60% of the  $^{21}\text{Ne}_c$  is released below  $350^\circ\text{C}$ , and above  $600^\circ\text{C}$  less than 5% of the  $^{21}\text{Ne}_c$  remain in the quartz. The stepwise extraction data can be used to estimate the activation energy for diffusion of Ne from quartz. The data  $\leq 350^\circ\text{C}$  yield activation energies of  $\approx 100\text{kJ mol}^{-1} \text{K}^{-1}$ .

$^{26}\text{Al}$  concentrations and  $P_{26}$  rates have previously been determined for quartz separates of Sierra Nevada rocks which were brought to the surface by glacial scouring during the Tioga period of the last ice age  $\approx 11000$  years ago [5]. Erosion during the last 11000 years has been less than a millimeter as evidenced by still visible glacial polish. Therefore, these samples appeared well suited for a calibration of the production rate ratio  $P_{21}/P_{26}$ . For this work, we selected two of the previously studied quartz separates W86-8, and W86-12 from 3556m and 2452m altitudes, respectively.

As it turned out, the Ne inventory in these samples is much more complicated than in the Antarctic quartz samples. Furthermore, only about  $2 \times 10^6$  atoms/g  $^{21}\text{Ne}_c$  are expected due to the short exposure age. The following Ne components have been identified in addition to spallation Ne:

1) Ne produced by the reaction  $^{18}\text{O}(\alpha, n)^{21}\text{Ne}$ . Fig. 2 shows the release pattern of excess  $^{21}\text{Ne}$  from a density separated grain-size fraction of sample W86-12. The  $(\alpha, n)$ -produced component is mainly released below  $600^\circ\text{C}$ .

2) Trapped Ne is predominantly released between  $400$ - $800^\circ\text{C}$ . The higher release temperature of  $\text{Ne}_{tr}$ , if compared to  $^{21}\text{Ne}_c$ , indicates that it is at least in part located in different sites. This is supported by the fact that  $^{20}\text{Ne}$  abundances show considerable variation in different splits of the same sample. Trapped Ne in density separated 38-90 and 90-125  $\mu\text{m}$  grain-size fractions of sample W86-12 are lower by about a factor of 4. However, all other interfering components are still present. Trapped Ne in sample W86-12 is slightly lighter than atmospheric Ne, while  $\text{Ne}_{tr}$  in the density separated grain size fractions is within error limits consistent with an atmospheric composition.

3) At high temperatures ( $>800^\circ\text{C}$ ) an additional Ne component ( $\text{Ne}_{HT}$ ) is released that resembles cosmic-ray produced Ne in its isotopic composition. Based on the release profile as well as the amounts of excess  $^{21}\text{Ne}$ , this component cannot possibly have been produced *in-situ* by cosmic-rays during the last 11000 a. Alternate

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production mechanisms are  $(\alpha, n)$ -reactions in minerals with an O/F ratio of  $\approx 200$ , or reactions by fission neutrons. Using activation energies and diffusion constants estimated from the high temperature release, we corrected this component at lower temperatures. These corrections are small as can be seen from Fig. 2.

The release patterns obtained from ALH85-4 as well as W86-12 (Fig. 2), show that more than 95% of  $^{21}\text{Ne}_c$  is released below  $600^\circ\text{C}$ . The total  $^{21}\text{Ne}$  excess released below  $600^\circ\text{C}$  in two splits of sample W86-8 is  $2.1 \pm 0.4$  and  $3.4 \pm 0.4$  atoms, respectively. Therefore, the lower of these values yields an upper limit for the amount of cosmic-ray produced  $^{21}\text{Ne}$  in sample W86-8. The corresponding production rate ratio is  $P_{21}/P_{26} \leq 0.63 \pm 0.1$ . Corrections for contributions from the high-temperature component are smaller than the stated error. Alternatively,  $^{21}\text{Ne}_c$  in the density separated 38-90 and 90-125  $\mu\text{m}$  grain-size fractions of sample W86-12 was determined from the Ne released below  $600^\circ\text{C}$  assuming that Ne in the two grain size fractions is a three component mixture of trapped, nucleogenic, and spallation Ne. Unfortunately, the calculated production rate ratio  $P_{21}/P_{26}$  depends critically on the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio of the trapped component. Based on Ne three isotope diagrams of all splits from sample W86-12, we conclude that the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio of the trapped component cannot be larger than atmospheric. The minimum value for  $P_{21}/P_{26}$  is  $0.70 \pm 0.15$ , and is obtained for a trapped component of atmospheric composition.

Our  $P_{21}/P_{26}$  value is considerably larger than theoretical estimates of 0.25-0.5 for Ne produced from Si at large shielding depths [6, Reedy priv. comm.]. On the other hand, in the center of an ordinary chondrite with 65 cm radius, Michel *et al.* [3] calculate a production rate ratio from Si  $P_{21}/P_{26} = 0.68$ .

**References :** [1] C.P. Kohl and K. Nishiizumi (1992) *Geochim. Cosmochim. Acta* 56, 3583-3587. [2] K. Nishiizumi, C.P. Kohl, J.R. Arnold, J. Klein, D. Fink, and R. Middleton (1991) *Earth Planet. Sci. Lett.* 104, 440-454. [3] R. Michel, P. Dragovitsch, P. Cloth, G. Dagge, and D. Filges (1991) *Meteoritics* 26, 221-242. [4] C.M. Hohenberg, K. Marti, F.A. Podosek, R.C. Reedy, and J.R. Shirk (1978) *Proc. Lunar Planet. Sci. Conf.* 9, 2311-2344. [5] K. Nishiizumi, E.L. Winterer, C.P. Kohl, J. Klein, R. Middleton, D. Lal, and J.R. Arnold (1989) *J. Geophys. Res.* 94, 17,907-17,915. [6] D. Lal (1988) *Ann. Rev. Earth Planet. Sci.* 16, 355-388.

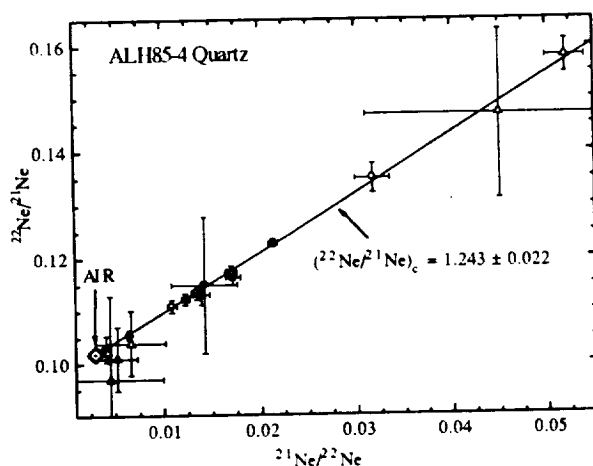


Fig. 1: Ne three-isotope diagram of data from ALH85-4. Error bars correspond to 95% confidence levels. All data lie on a straight line passing through the "AIR" point (atmospheric composition). From the best-fit line we calculate  $(^{22}\text{Ne}/^{21}\text{Ne})_c = 1.243 \pm 0.022$ .

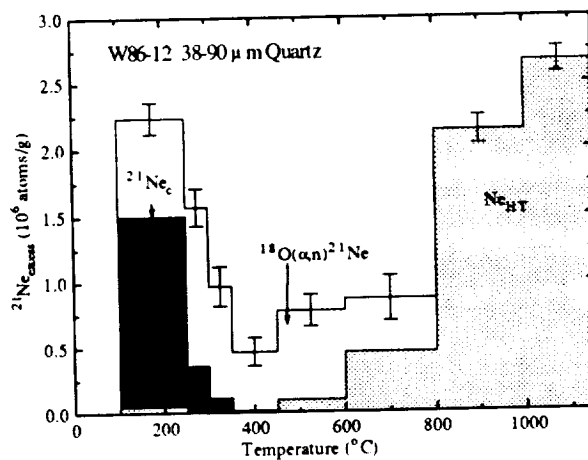


Fig. 2: Release pattern of excess  $^{21}\text{Ne}$  from the density separated 38-90  $\mu\text{m}$  grain-size fraction of sample W86-12. At low temperatures, excess  $^{21}\text{Ne}_c$  is of nucleogenic and cosmogenic origin. At high temperatures the release is dominated by  $^{21}\text{Ne}_{\text{HT}}$  (see text). From the high temperature release we estimate an activation energy of  $\approx 115 \text{ kJ mol}^{-1} \text{ K}^{-1}$  for this component. Due to the high activation energy, the contribution at low temperature is small.